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High Fluence, Multi-pulse Laser Surface Damage: absorbers, mechanisms and mitigation

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High Fluence, Multi-pulse Laser Surface Damage: absorbers, mechanisms and mitigation

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All optical systems designed to guide high photon fluxes are limited by optical damage and optical stress induced degradation to key optical components including lenses, frequency conversion crystals, anti-reflection coatings and dielectric multilayer mirrors. These systems include continuous wave (CW) and Giga-shot high average power (HAP) systems for laser machining and DoD applications, and systems for laser driven inertial confinement fusion (ICF) and inertial fusion energy (IFE). Optical degradation and damage begin with near surface defect absorption of sub band-gap light; these effects appear beginning in the near IR (1eV) and become progressively worse for light in the near UV (3 to 4eV). At high enough per pulse fluences (ϕ), localized energy deposition is sufficient to create damage sites through explosive ejection of material; this process, optical damage initiation, results in micron sized surface pits which can grow exponentially upon exposure to additional pulses.

Much progress has been made understanding and reducing optical damage in silica. In previous work, the optical and electronic properties of precursors from surface flaws were studied, and defect layers on fractures surfaces were identified as fluence limiting precursors; subsequently, chemical etch processes (AMP –Advanced Mitigation Process) were developed which greatly suppressed damage from these fractures. Figure 1 shows the expected number of initiations for a full NIF-sized silica lens after AMP processing versus per pulse fluence ϕ_p along with fluence distributions for ICF systems such as NIF, IFE systems and high average power quasi-CW systems. Total lifetime fluences Φ_{LT} (shots multiplied by per pulse fluence), in these systems are shown as arrows. Initiation from fractures (gray curve) has been greatly suppressed by AMP. However, even fracture-free surfaces damage at fluences only slightly higher than the fractures. These "high fluence (ϕ)" precursors (black line) have a strong threshold, far below the intrinsic threshold of the bulk material, and ultimately limit the maximum fluence an optic can see. The damage behavior for silica shown in Figure 1 is typical of all optical materials, though the precursors may be different. One important goal of this project was to determine the nature of these high ϕ precursors, identify their sources and find practical mitigations for them.

At much lower ϕ, photochemical changes can degrade optical performance in less catastrophic ways including loss of transmission (quasi-CW HAP systems which see large Φ_{LT}). Optics for HAP systems will see billions of pulses below optical damage "thresholds" and can potentially degrade or damage. Although they are designed to operate well below damage thresholds, modulation and high ϕ damage can also be a problem for these lasers. Little is known about the lifetime of optics under these conditions, as it is very difficult to design systems capable to of this many high fluence shots, and the long times required for comprehensive sample testing are prohibitive. Prior to this work, the mechanisms responsible for optical degradation and its behavior as a function of laser variables up to Giga-shot levels was unknown.

The goals of this proposal were (1) to develop a scientific understanding of the mechanisms and precursors which affect high fluence optical damage in silica and to develop practical methods for their mitigation, and (2) to explore how optical stress-induced degradation scales with pulse fluence and intensity in order to clarify the mechanisms responsible for degradation and to develop practical schemes for accelerated testing out to Giga-shot levels.

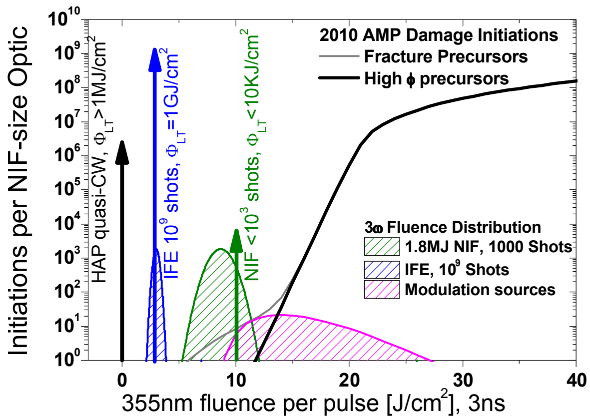

Figure 1: Initiations on a large aperture (1100cm^2) AMPed silica optic as a function of 3ns, 355nm pulse fluence: fracture-related precursors (gray), high fluence precursors (black). Also shown are typical fluence distributions due to beam contrast for three systems which transport high lifetime fluences Φ_{LT}; Φ_{LT} is indicated, and lifetime systems shots shown as arrows.

This project was organized into the following tasks:

1. Perform experiments and develop models to understand the physics of high ϕ precursor absorption and damage initiation
2. Develop techniques to identify high ϕ precursors and the process steps which introduce them to fracture-free optical glasses
3. Develop processes to reduce high fluence damage initiation
4. Explore methods for accelerated optical stress testing, and clarify the mechanisms responsible for stress-related degradation:

All tasks were successfully completed. Key results from the first task are summarized in reference [1] below. A native surface precursor can absorb sub band-gap light and initiate a process which leads to catastrophic damage many micrometers deep with prominent fracture networks. However, previous model precursor systems designed to study initiation experimentally were not able to clearly reproduce these damage events. In this study, we created artificial absorbers on fused silica substrates to investigate precursor properties critical for native surface damage initiation. Thin optically absorbing films of different materials were deposited on silica surfaces and then damage tested and characterized. We demonstrated that strong interfacial adhesion strength between absorbers and silica is crucial for the launch of absorption front and subsequent damage initiation. Simulations using the absorption-front model were performed and agree qualitatively with experimental results. In addition, reference [2] completes a study correlating quasi-continuum (QC) photoluminescence (PL) with the electronic and optical properties of damage precursors. The source of the PL is not attributable to any known silica point defect. The primary features of QC-PL include broad excitation and emission spectra, a broad distribution of photoluminescence lifetimes from 20 ps to 5 ns, continuous shifts in PL lifetime distributions with respect to emission wavelength, and a propensity to photo-bleach and photo-brighten. We found similar photoluminescence characteristics in surface flaws of other optical materials, including CaF_2, DKDP and quartz. Based on the commonality of the features in different optical materials and the proximity of QC-PL to surfaces, we suggest that these properties arise from interactions associated with high densities of defects, rather than a distribution over a large number of types of defects and is likely found in a wide variety of structures from nano-scale composites to bulk structures as well as in both broad and narrow band materials from dielectrics to semiconductors. Reference [3] presents a study of the thermal annealing properties of damage precursors.

Results from tasks (2) and (3) are summarized in references [4]-[8]. First, reference [4] describes a study of the energy distribution of high fluence precursors performed in order to help identify and classify potential sources of high fluence precursors. The density of damage precursors at low fluence had been measured using large beams (1-3 cm); higher fluences cannot be measured easily since the high density of resulting damage initiation sites results in clumping of damage sites. Here, we developed automated experiments and analysis that allow us to damage test thousands of sites with small beams (10-30 μm), and automatically image the test sites to determine if laser damage occurred. We developed a rigorous connection between these small beam damage test results of damage probability versus laser pulse energy and the large beam damage results of damage precursor densities versus fluence. We used a fitting model that accounts for the fluence distribution within the beam to determine the density of laser damage precursors. We also developed a fitting procedure using Levenberg-Marquardt minimization for the maximum likelihood estimator of the binomial distribution. We found that for uncoated and coated fused silica samples, the distribution of precursors does not increase more rapidly at very high fluences, up to 150 J/cm^2.

Reference [5] describes the experiments we conducted to identify the source of high fluence precursors on silica optics and methods to mitigate them. This work presents data supporting the hypothesis that high fluence precursors (e.g. UV fluences >10 J/cm^2 for pulses 5 ns in duration) on silica optical surfaces are generated as precipitates during chemical processing steps such as cleaning, etching, rinsing. We have shown that a variety of ionic species, when precipitated on optical surfaces over a wide size scale, can lead to optical damage. These precipitates need not be composed of bulk optical absorbers at the wavelengths of interest, and in fact the damage behavior appears to be nearly independent of composition. We provide evidence that suggests that the presence of such precipitates at size scales below that which can be reliably observed by optical or even electron microscopy represent a significant barrier to the fabrication of UV optics for high fluence applications. Finally, by working to exclude reagent and process contamination and to minimize precipitation during chemical processing operations, we have demonstrated the production of silica optics with extraordinarily low damage densities which show saturated precursor densities of ≈200 cm^{-2} on test samples, reducing damage density in silica at high fluence by 300 times while shifting the fluence onset of observable damage by about 7 J/cm^2.

In reference [6], we also showed that trace impurities in ultrapure water used to process fused silica optics may be responsible for the formation of carbonaceous deposits. We used surrogate materials to show that organic compounds precipitated onto fused silica surfaces form discrete damage precursors. Combined with a standard etching process, solvent-free oxidative decomposition using oxygen plasma or high-temperature thermal treatments in air reduced the total density of damage precursors to as low as <50 cm^{-2}. This corresponds to a reduction in damage density of 2000 times and a threshold shift of almost 10J/cm^2, nearly 70% of a 1.8MJ NIF shot. Finally, we showed that inorganic compounds are more likely to cause damage when they are tightly adhered to a surface as per the work in task 1 reference [1], which may explain why high-temperature thermal treatments have been historically unsuccessful at removing extrinsic damage precursors from fused silica. Reference [7] combines this work with previous studies and presents an overall summary of silica laser damage mechanisms, precursors and their mitigation.

Task (4) explores the physics of Giga-pulse degradation for UV light. Key results of this study are presented in reference [8]. As applications of lasers demand higher average powers, higher repetition rates, and longer operation times, optics will need to perform well under unprecedented conditions. We investigated the optical degradation of fused silica surfaces at 351 nm for up to one billion pulses with pulse fluences up to $12 J/cm^2$. The central result is that the transmission loss from defect generation is a function of the pulse intensity, I_p, and total integrated fluence, φ_T, and is influenced by atmospheric conditions. In 10^{-6} Torr vacuum, at low I_p, a transmission loss is observed that increases monotonically as a function of number of pulses. As the pulse intensity increases above 13 MW/cm^2, the observed transmission losses decrease, and are not measureable for 130 MW/cm^2. A physical model which supports the experimental data is presented to describe the suppression of transmission loss at high pulse intensity. Similar phenomena are observed in anti-reflective sol-gel coated optics. Absorption, not scattering, is the primary mechanism leading to transmission loss. In 2.5 Torr air, no transmission loss was detected under any pulse intensity used. We found that the absorption layer that leads to transmission loss is less than 1 nm in thickness, and it results from a laser-activated chemical process involving photo-reduction of silica within a few monolayers of the surface. The competition between photo-reduction and photo-oxidation explains the measured data: transmission loss is reduced when either the light intensity or the O_2 concentration is high. We expect processes similar to these to occur in other optical materials for high average power applications.

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